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Microwave Dielectric Properties of (1-*x*)$Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$–*x*$NdAlO_3$ Ceramics

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Abstract: This study presents the microwave dielectric properties calculation of $(1-x)Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$–$xNdAlO_3$ ceramics where x denotes the volume molar fraction. From X-ray diffraction results, the solid solution limit is calculated to be about 0.76, where it forms a single $BaNd_2Ti_4O_{12}$ phase in Region I ($0 \le x \le 0.76$), and both $BaNd_2Ti_4O_{12}$ and $NdAlO_3$ coexist in Region II ($0.76 < x < 1$). The solid solution limit is confirmed by independently calculating it from the dielectric constant data. There is less than 4 % deviation between the measured dielectric constant (ε_r) and the one calculated from the Maxwell-Wagner formula. The total quality factor (Q) remains almost constant in Region I and increases rapidly with the volume molar fraction of $NdAlO_3$ in Region II. The measured $Q \times f$, where f is the resonant frequency, is also consistent with the calculated value in both regions. The temperature coefficient at the resonant frequency is -1.4 ppm/^{o}C, which agrees well with the calculated value of 0 ppm/^{o}C. In addition, we observed a close correlation between the bulk density and the phase evolution.

Keywords: Microwave Dielectric Properties; Maxwell-Wagner Formula; Solid Solution Limit.

I. Introduction

Microwave dielectric ceramics play an increasingly important role in a wide range of microwave frequency devices. For example, resonators made from these ceramics provide high dielectric constants (ε_r), high quality factors (Q), and near-zero temperature coefficients (τ_f) at the resonant frequencies.[1-5] The size of devices made from ceramics is proportional to $\varepsilon_r^{-1/2}$, so a higher dielectric constant will decrease the device footprint.[1-5] The

high quality factor is desirable because it allows for higher precision frequency selection.[1,3,6] Furthermore, the stability of a device is characterized by temperature coefficient of resonant frequency:

$$\tau_f = \frac{f_{t_2} - f_{t_1}}{f_{t_1} \times (t_2 - t_1)} \times 10^6 \quad (1)$$

in which the units of τ_f are ppm/^{o}C, f_{t1} is the resonant frequency at the room temperature (t_1) and f_{t2} is the resonant frequency at t_2 = 85 ^{o}C. [1,3,7] The most stable situation of τ_f = 0 ppm/^{o}C is usually required.[1] However, most ceramics inherently possess non-zero temperature coefficients.[1] Thus, wide studies in recent decades of adjusting temperature coefficients to zero have been performed in high dielectric constant ceramics, for example, $CaTiO_3$, $BiVO_4$ and $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$ based ceramics.[1,7-13]

One of the most effective and widely used methods for obtaining a zero temperature coefficient is to combine two phases with τ_f of opposite signs via

$$\tau_f = (1-x)\tau_{f1} + x\tau_{f2} \quad (2)$$

where x denotes volume molar ratios and τ_{f1} and τ_{f2} have opposite signs.[4,5,14] Most of the reported τ_f results fit well with Equation (2) while the variation of the dielectric constant and quality factor always depends on whether the solid solution is formed or not.[11-14] If two phases coexist, the $Q \times f$ can usually be easily predicted as in Refs [13], [15] and [16].[13,15,16] When a single phase solid solutions is formed through the linear combination of Equation (2), the theoretical calculations of ε_r and $Q \times f$ deviate largely from experimental results, for example, in Refs [4], [13] and [14].[4,13,14]

This study focuses on modeling the dielectric constants and quality factors of ceramic mixtures, i.e. $(1-x)Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$–$xNdAlO_3$ ceramics. The compound is a single- phase solid solution when $0 < x \leq 0.76$ and a two- phase system when $0.76 \leq x < 1$.

II. Experimental procedure

The compounds in this study, $Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$ (BNCNTO) and $NdAlO_3$ (NAO) are synthesized respectively according to methods in Refs [17] and [18] respectively. High-purity (≥ 99%) materials are used: $BaCO_3$ (Xilong Group, China), Nd_2O_3 (Jichang Rare Earth Co., Ltd., China), TiO_2 (Xian Tao Chemical Reagent Co., Ltd., China), Nb_2O_5 (Ke Long Chemical Reagent Co., Ltd., China), Cr_2O_3 (Ke Long Chemical Reagent Co., Ltd., China), Al_2O_3 (Betterwear New Matrial Co., Ltd., China). Then, BNCNTO and NAO are mixed, according to (1-*x*)BNCNTO–*x*NAO, with *x* = 0, 0.2, 0.4, 0.6, 0.8 where the data for *x* = 1 will be cited from [18]. The mixtures are ball-milled in nylon jars with zirconia balls and deionized water for 12 hours. They are then dried at 120 ^{o}C for 24 hours. Cylindrical 14.5 mm (diameter) × 6.5 mm (thickness) samples are formed with a 5 wt% polyvinyl alcohol (PVA) solution by applying of 250 kg/cm^2 pressure for 30 seconds. The samples are heated at 600ºC for 2 hours to remove the PVA binder and then they are sintered at 1350 – 1450 ºC for 2 hours. Finally, cylinders are left to naturally cool to room temperature.

The bulk densities of the sintered samples are measured by the Archimedes' method. The phase of the sintered samples is identified by the powder X-ray diffraction (XRD) using Cu K alpha radiation (40 kV/250 mA, wavelength (λ) = 1.54062 Å, Philips x'pert Pro MPD, PANalytical, the Netherlands). The surface microstructure of the ceramics is characterized by scanning electron microscopy (SEM, FEI Inspect F). The dielectric characteristics are measured by the Hakki-Coleman dielectric resonator method in the TE_{011} mode using a network analyzer (Agilent Technologies E5071C, Agilent Technologies, Singapore) at the frequencies around 4 GHz. The temperature coefficients at the resonant frequency of the sintered samples are calculated through Equation (1).

III. Results and discussion

For X-ray diffraction results, Ref. [19] proved that the sintering temperature will not influence the phase peak position and that the peak intensity will be maximized when samples sintered at the optimal temperature.[19] In the bulk density part, we will show that the optimal temperature is 1425 °C. Thus, in Figure 1(a), we show X-ray diffraction patterns of (1-*x*)BNCNTO–*x*NAO ceramics sintered at 1425 °C. When $0 \le x \le 0.6$, the compound is in a single $BaNd_2Ti_4O_{12}$ phase and there is no evidence of the $NdAlO_3$ phase.[20] We define the relative 2ϑ shift D_r as $D_r = D_{x\neq 0} - D_{x=0}$, in which $D_{x\neq 0}$ and $D_{x=0}$ are the (511) peak positions of samples with and without the $NdAlO_3$ addition. In Figure 1(b), the relative 2ϑ shift (circular dots) shows a growing trend from $x = 0$ to 0.6. In other words, the (511) peak position shifts towards a larger angle 2ϑ. An increase in D_r indicates a decrease in the unit cell volume since, according to Bragg's law, $2d\sin\theta = \lambda$, where d is lattice plane separation, ϑ could be obtained by 2ϑ degree in XRD, and λ is the fixed incident wavelength of 1.54062 Å.[21] It is possible to have a phase where $NdAlO_3$ dissolves into $BaNd_2Ti_4O_{12}$ phase because: 1) R_{Nd}^{3+} (0.0983 nm) ≤ R_{Ba}^{2+} (0.135 nm) and R_{Al}^{3+} (0.0535 nm) ≤ R_{Ti}^{4+} (0.0605 nm), 2) Ref. [3] has categorized both materials as the perovskite related materials, and 3) only one phase exists.[3,22] From $x = 0.6$ to $x = 0.8$, D_r appears to saturate. Therefore, we here initially conclude that a solid solution between $BaNd_2Ti_4O_{12}$ and $NdAlO_3$ is formed and Nd^{3+} and Al^{3+} dissolve into A and B sites respectively for $0 < x \le 0.6$.[1] The solid solution limit (m) should be between 0.6 and 0.8.

The formation of the solid solution can be confirmed by the variation in the peak intensities. In Figure 1(a), the (511) peak of the $BaNd_2Ti_4O_{12}$ phase is around 32°.[20] The main peak of the $NdAlO_3$ phase and the (231) peak of the $BaNd_2Ti_4O_{12}$ phase are both around 33°.[20,23] Tseng *et al.* report a simple way to estimate the phase content ratio by comparing the peak intensities.[24] We define the peak intensity ratio (I_r) through

$I_r = I_{(231)} / I_{(511)}$, where $I_{(231)}$ and $I_{(511)}$ are the (231) and (511) peak intensities, respectively. In Figure 1(b), I_r remains constant of 0.4 in $0 \le x \le 0.6$ and jumps to 1.1 for $x = 0.8$, indicating the formation of the solid state solution. From the peak intensity ratio, we can obtain the solid solution limit. When $x = m$, the volume molar ratio of $BaNd_2Ti_4O_{12}$ and $NdAlO_3$ are 1-m and m. When $x = 0.8$, the volume molar ratio of $BaNd_2Ti_4O_{12}$ phase is 0.2. The amount of $NdAlO_3$ that can dissolve into $BaNd_2Ti_4O_{12}$ is $0.2m/(1-m)$ and $(0.8-m)/(1-m)$ $NdAlO_3$ is left. The solid solution limit $m \approx 0.76$ can then be extracted from $[(0.8-m)/(1-m)]/0.2 = 1.1$. The solid solution limit will be independently calculated a second time below from the dielectric constant data. So far, we can separate the volume molar ratio into two regions: Region I, $0 < x \le 0.76$, where the solid solution is formed and Region II, $0.76 \le x < 1$, where two phases co-exist.

The bulk density data of (1-x)BNCNTO – xNAO are shown in the inset of Figure 2. First, for each value of x, the sample density increases as the sintering temperature increases from 1400 ^{o}C to 1425 ^{o}C and then the density decreases when the temperature reaches 1450 ^{o}C. This indicates that the ceramic will be most compact when sintered at 1425 ^{o}C. Secondly, when sintered at 1425 ^{o}C, the bulk density increases with x. When x varies from 0 to 0.6, the density rises very slowly from 5.6 g/cm^3 to 5.7 g/cm^3, which is very close to the density of the $BaNd_2Ti_4O_{12}$ phase.[9,17,18] The growing density may originate from the compact microstructure shown in Figure 3 (a)-(d). Details of the SEM result will be discussed later. However, until x = 0.6, the bulk density is still much lower than the density of the $NdAlO_3$ phase (ρ_{NA} ~ 6.9 g/cm^3), which confirms the formation of the $BaNd_2Ti_4O_{12}$ solid solution between the two phases.[18] When $x \ge 0.8$, the density increases rapidly from 5.8 g/cm^3 to 6.9 g/cm^3, which is due to the presence of the $NdAlO_3$ phase (ρ_{NA} ~ 6.9 g/cm^3). The changes in the density behavior in the two regions indicate the close correlation between the bulk density and the phase evolution.

Microstructures of samples sintered at 1425 ºC are shown in Fig. 3. All compositions show rod-like grains, which are typically observed in $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$ ceramics.[2,7,24,25,26] The surface of the sample without $NdAlO_3$ dopant is comprised of large- and small- size grains. As *x* increases from 0 to 0.6, more and more grains grow longer and more robust, with more homogeneous morphology.[9] This result is consistent with that of the increasing bulk density: since the ceramic keeps a single $BaNd_2Ti_4O_{12}$ phase, the total density is similar to the $BaNd_2Ti_4O_{12}$ phase; the microstructure of the ceramics become more and more compact, so the density increases.

When $x = 0.8$, grains in small sizes appear again. Given that the existence of the $NdAlO_3$ phase has been proved by the XRD results, while Ref. [29] has also demonstrated that its grain size is about 1 μm, we can conclude that these small grains are $NdAlO_3$ grains. To further confirm this, we perform the energy dispersive X-ray spectroscopy (EDS) and the results are shown in Table 1. By adding the number of Cr and Nb into Ti site, the element ratio of Ba : Nd : (Ti,Cr,Nb) : O of A and B are estimated to be 1 : 2 : 4 : 12, both of which are $BaNd_2Ti_4O_{12}$ phase. In spot C, if we ignore the minor amount of Ba and Ti, the element ratio of Nd : Al : O is around 1 : 1 : 3, which is the $NdAlO_3$ phase. In spot D, if it is treated as a solid solution, the element ratio of Ba : Nd : (Ti,Al,Cr,Nb) : O is about a sum of 1 : 2 : 4 : 12 and 1 : 1 : 3. As a result, although samples of $x = 0$ and $x = 0.8$ both contain small-size grains (B and C), they are totally different phases, i.e. $BaNd_2Ti_4O_{12}$ and $NdAlO_3$, respectively. These results correspond to the fact that: 1) two phases co-exist as is proved in the XRD results; 2) bulk density rapidly increases because of the existence of the $NdAlO_3$ phase for $x \geq 0.8$.

Figure 2 depicts the dielectric constant of (1-*x*)BNCNTO–*x*NAO ceramics sintered at different temperatures. For each *x* value, the dielectric constant is sustainably growing as the sintering temperature increases from 1400 ^{o}C to 1425 ^{o}C, while the dielectric constant shows a decreases at 1450 ^{o}C. A similar trace between

dielectric constant and temperature, compared to bulk density, confirms the claim that the bulk density can predict the variation of dielectric constant versus temperature.[30] A higher bulk density indicates a higher dielectric constant, because of lower porisity.[30] The results of dielectric constant versus temperature are in good agreement with the density and SEM results.

Then for different compositions sintered at 1425 ^{o}C, the dielectric constant keeps decreasing. In general, the dielectric constant can be estimated by the Maxwell-Wagner mixing rule:

$$\varepsilon_{mix}^{k} = (1-x)\varepsilon_{1}^{k} + x\varepsilon_{2}^{k} \quad (3)$$

where ε_{mix} is the dielectric constant of the mixed ceramic, ε_1 and ε_2 are the dielectric constant of two original ceramics, and k is the constant for different models.[10,13-16] In the following, through Equation (3), a new k value will be determined for the solid solution for $0 \le x \le 0.6$ and use the conventional $k = 1$ to fit the data for $0.8 \le x \le 1$.[13-16] From the analysis of XRD result, the solid solution is about 0.76, in the following we will calculate it independently from the dielectric constant data. After that, we will calculate the relative deviation between the calculated and measured data to confirm its effectiveness.

For $0 \le x \le 0.6$, the solid solution is formed and the reported k values are invalid.[13,14] This is because conventional models presume the co-existence of two phases with parallel or series distribution.[15] Mathematically, from Equation (3), $k = 1$ is a linear relation and its data lies closer to experimental results than other k values smaller than 1. All experimental data exhibit a nonlinear trend. Therefore, by applying formula and continuously adjusting k value larger than unity, it finds that when $k = 4$ the calculated results match measured data perfectly, as shown in Figure 5. For $x = 0.8$, we have observed the $NdAlO_3$ phase. E. Kim et *al.* have reported the effectiveness of the parallel Maxwell-Wagner equation, $k = 1$, for the multiple phase

dielectric constant calculation.[10] As a result, once the solid solution limit is determined, we can calculate the effective dielectric constant of the solid solution, and thus estimate the dielectric constant of $0.8 \le x \le 1$.

We here provide the explicit steps for the calculation of solid solution limit. The solid solution limit should be $0.6 < m < 0.8$. When $x = m$, volume fractions of $NdAlO_3$ and $BaNd_2Ti_4O_{12}$ are m and $1- m$. It follows Equation (3):

$$\varepsilon_m^4 = (1-m)\varepsilon_{BNCNTO}{}^4 + m\varepsilon_{NAO}{}^4 \quad (4)$$

where, ε_m is the dielectric constant when $x = m$, to be determined. For $x = 0.8$, two phases co-exist, also obeying Equation (3), with $k = 1$. $0.2m/(1-m)$ $NdAlO_3$ dissolves into $0.2/(1-m)$ $BaNd_2Ti_4O_{12}$ phase while $0.8 – 0.2m/(1-m)$ $NdAlO_3$ is remained. Then we can obtain:

$$\varepsilon_r|_{x=0.8} = (0.8 - \frac{0.2m}{1-m})\varepsilon_{NAO} + \frac{0.2}{1-m}\varepsilon_m \quad (5)$$

where $\varepsilon_{r|x=0.8}$ is an experimental result. Based on the data of $\varepsilon_{r|x=0.8}$, ε_{BNCNTO}, ε_{NAO}, we depict traces of Equation (4) and (5) in Figure 4 and obtain solid solution around 0.76 and $\varepsilon_m = 56.34$. The value of $m = 0.76$ is the same value as what we have calculated from the XRD data. We find that around 3.17 times $NdAlO_3$ phase can be dissolved into the $BaNd_2Ti_4O_{12}$ phase. The relative deviation, $\frac{\varepsilon_{calculated} - \varepsilon_{measured}}{\varepsilon_{measured}} \times 100\%$, between the calculated ($\varepsilon_{calculated}$) and experimental ($\varepsilon_{measured}$) dielectric constants, is calculated to confirm the effectiveness of our results. In the inset of Figure 4, $0 \le x \le 1$, all relative deviations are less than 4 %. Up to now, the effectiveness of $m = 0.76$ has been ensured. Thus, we can conclude that Maxwell-Wagner equation matches very well with experimental results, as well as the new value of $k = 4$ is effective for the solid solution.

Besides the dielectric constant, quality factor is another important property parameter of ceramics. The quality factor of the mixed ceramic is usually related to the dielectric losses (*tanδ*) of individual phases, *tanδ* ~ 1/*Q*, which can be added linearly:

$$Q_{mix}^{-1} = (1-x)Q_1^{-1} + xQ_2^{-1} \quad (6)$$

where Q_{mix} is the quality factor of the mixed ceramic, while Q_1 and Q_2 are the quality factors of two original ceramics.[4,5] Figure 5 displays the measured quality factor (*Q*) of (1-*x*)BNCNTO–*x*NAO ceramics. When $0 < x \leq 0.6$, it forms a $BaNd_2Ti_4O_{12}$ solid solution, as concluded in the dielectric constant section. Thus, it is reasonable to set *x* as zero and the total quality factor is dominated by the $BaNd_2Ti_4O_{12}$ phase. Prior reports on $Ba_{6-3x}Nd_{8+2x}Ti_{18}O_{54}$ ceramics show that their quality factors are mostly in the vicinity of 3000 (*f* ~ 4 GHz).[7-9,25-27] Thus, quality factors of around 3000, far less than the quality factor of the $NdAlO_3$ phase, is reasonable.[18] In addition, by performing attentive observation, we can find that quality factor slightly increases for $0 \leq x \leq 0.6$. It has been reported that a larger grain size would result in less grain boundaries per unit volume, which leads to less two dimensional defects breaking symmetry periodicity.[31] From Figure 3, we obtain larger and larger grain size, and it is logical to observe the increasing quality factor.[30-32]

For *x* = 0.8, quality factor is however higher than 3200. According to Equation (6), a phase ratio of roughly 1:1 ($x \approx 0.5$ in Equation (6)) makes the total quality factor mainly determined by the low *Q* phase and the slight increase is attributed to the existence of the $NdAlO_3$ phase. Consequently, we conclude that the quality factor is controlled by microstructure and the phase constitution.

In practice, more attention should be paid to *Q×f* (multiplication between the quality factor and resonant frequency), as shown in Figure 6. Since these cylinders are essentially Fabry-Perot resonators, the resonant frequency is proportional to the reciprocal of the square root of dielectric constant:

$$f \propto \frac{1}{L\sqrt{\varepsilon_{mix}}} \quad (7)$$

where *f* is the resonant frequency, *L* is the diameter of the cylinder and ε_{mix} is the dielectric constant defined as in Equation (3).[1,3,33,34] In Figure 4, the dielectric constant keeps decreasing, which results in an increasing resonant frequency, if the size of all samples are assumed to be fixed for simplicity. In Region I, the *Q×f* value of each composition, which is determined by the $BaNd_2Ti_4O_{12}$ phase, can be calculate through multiplying *Q×f* of the $BaNd_2Ti_4O_{12}$ phase by the relative resonant frequency shift (f_{shift}) $Q \times f = Q_{x=0} \times f_{x=0} \times f_{shift}$. The relative resonant frequency shift (f_{shift}) is defined as, $f_{x\neq0}/f_{x=0}$, where $f_{x\neq0}$ is the resonant frequency for $x \neq 0$ and $f_{x=0}$ is the resonant frequency for $x = 0$, as shown in Figure 5. The increasing relative resonant frequency shift (f_{shift}) results in increasing *Q×f* values.[18] In Figure 6, both measured and calculated *Q×f* are provided. In Region I, the difference between the calculated and measured *Q×f* is small, while in Region II, it starts to increase. We conclude that the *Q×f* in Region I is determined by the $BaNd_2Ti_4O_{12}$ phase, while the total *Q×f* in Region II is controlled by the phase constitution.

It has been always confirmed that the Equation (2) is precise enough to predict the τ_f variation without considering the phase state, for example, multiphase or solid solution, when two ceramics are mixing. Therefore, it is taken to calculate the τ_f value in the whole *x* range, where τ_f is the temperature coefficients of the mixture, τ_{f1} = 49 ppm/^{o}C and τ_{f2} = -33 ppm/^{o}C are temperature coefficients of two starting materials.[4,5,13,14] Figure 7 shows both measured and calculated temperature coefficient of resonant frequency values τ_f. The measured τ_f value exhibits a similarly linear decreasing trend as predicted by the theoretical results. As last, a

zero τ_f value at x = 0.6 is theoretically expected while the measured value is -1.4 ppm/^{o}C. It confirms the effectiveness of the Equation (2).

IV. Conclusions

In this study, we present a specific method to calculate microwave dielectric properties when two ceramics are mixing. The solid solution limit of 0.76 is calculated from the XRD result and verify it from the dielectric constant data. Two regions are separated by the solid solution limit, where in Region I, $0 \le x \le 0.76$, dielectric constant obeys the nonlinear Maxwell-Wagner mixing formula, quality factor is determined by the $BaNd_2Ti_4O_{12}$ phase and the microstructure; in Region II, dielectric constant obeys the linear Maxwell-Wagner mixing formula, quality factor is determined by the microstructure and the two phases. The relative deviation between the experimentally measured and the calculated dielectric constant is as low as 4 %. The $Q \times f$ is controlled by the microstructure, phase constitution and dielectric constant. We also have confirmed the method for realizing the goal of zero τ_f, by combining two ceramics with temperature coefficients of opposite signs. At last, an experimentally obtained τ_f = -1.4 ppm/^{o}C matches very well with the prediction of zero in Region I. We also observed the close correlation between the bulk density and the phase evolution.

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List of Figures

Figure 1(a). X-ray diffraction patterns of $(1-x)Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}–xNdAlO_3$ composite ceramics sintered at 1425 ^{o}C.

Figure 1(b). Selected part of peak position and intensity shift: left, peak intensity ratio, right, relative 2 theta shift.

Figure 2 Inset part: bulk density results of (1-*x*)$Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$–*x*$NdAlO_3$, sintered at 1400 ^{o}C, 1425 ^{o}C, 1450 ^{o}C; dielectric constant values of (1-*x*) $Ba_{3.75}Nd_{9.5}Cr_{0.5}Nb_{0.5}Ti_{17.5}O_{54}$ –*x*$NdAlO_3$, sintered at 1400 ^{o}C, 1425 ^{o}C and 1450 ^{o}C.

Figure 3 SEM photographs of (1-*x*)$Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$–*x*$NdAlO_3$ ceramics sintered at 1425 ºC: (a) *x* = 0, (b) *x* = 0.2, (c) *x* = 0.4, (d) *x* = 0.6, (e) *x* = 0.8.

Figure 4 Calculated dielectric constant results in two regions and measured dielectric constant data. Inset: relative deviation between calculated and measured results.

Figure 5 Left axis, measured quality factor results; right axis, relative resonant frequency shift.

Figure 6 Left axis, calculated *Q*×*f* value based on $BaNd_2Ti_4O_{12}$ phase in two regions; right axis, measured *Q*×*f* value of (1-*x*)$Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$–*x*$NdAlO_3$ ceramics.

Figure 7 Measured (circle dots) and calculated (line), through Equation (2), temperature coefficient of resonant frequency values τ_f.

Table 1. EDS data of (1-*x*)$Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$–*x*$NdAlO_3$ ceramics: A, B, C and D.

Table 1. EDS data of (1-*x*)$Ba_{3.75}Nd_{9.5}Cr_{0.25}Nb_{0.25}Ti_{17.5}O_{54}$–*x*$NdAlO_3$ ceramics: A, B, C and D.

Spot	Atom (%)						
	Ba	Nd	Ti	Cr	Nb	Al	O
A	4.41	11.81	20.61	0.29	0.31	-	62.57
B	4.59	11.61	20.23	0.31	0.35	-	62.91
C	-	20.58	-	-	-	20.61	58.81
D	3.58	12.93	17.1	0.23	0.21	3.61	62.34